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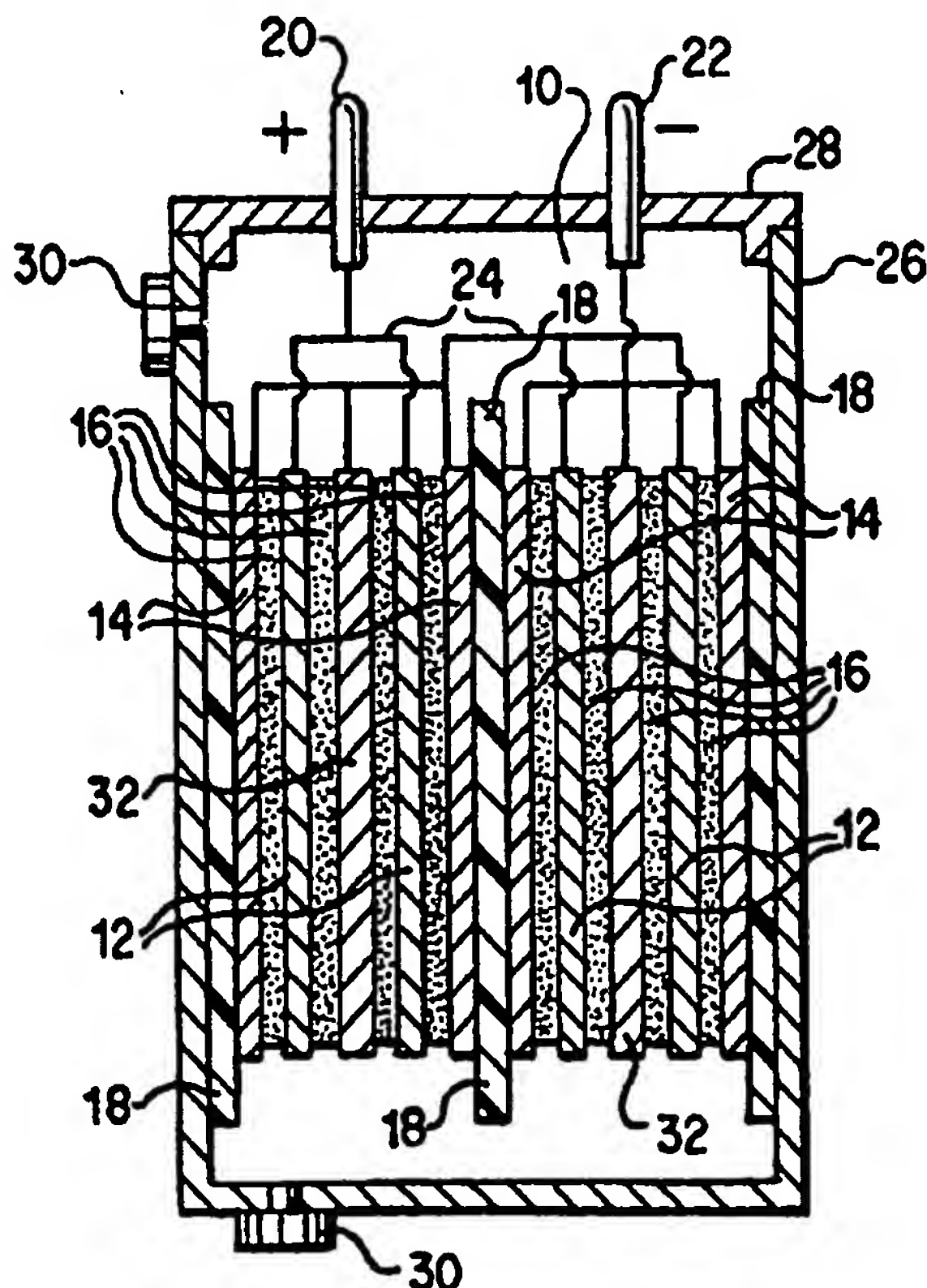
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(54) Title: WATER ACTIVATED CHEMICAL CURRENT SOURCE

(57) Abstract

A water activated chemical current source adapted for use with a water-based electrolyte includes a magnesium-based or aluminum-based anode, and a copper-base cathode comprising between about 55 wt % and about 85 wt % of copper hydroxide chloride, between about 5 wt % and about 20 wt % of sulfur, and between about 1 wt % and about 25 wt % of an electroconductive additive.



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WATER ACTIVATED CHEMICAL CURRENT SOURCE

Field of the Invention

The present invention generally relates to a water activated chemical current source and more specifically, relates to a water activated chemical current source comprising an anode of either magnesium or aluminum or their alloys, a cathode of a compound of copper hydroxide chloride, sulfur and an electroconductive additive, and a fresh or sea water electrolyte. The present invention further relates to a process of making such a water activated chemical current source.

Background of the Invention

The present invention relates to primary chemical current sources, i.e., water activated emergency current sources that can be used for power feeding of rescue floating means, radio beacons, buoys and meteorological probes:

A water activated current source, or a reserve battery, is a battery that is inert until an operation is performed which brings all the cell components into the proper state and location to become active. In water activated or electrolyte-activated batteries, the water or electrolyte component is not present during storage. It is added just before the cell is put into use. One of the more important design factors is to ensure that the electrolyte is delivered as quickly as possible at the time of activation, at the same time avoiding chemical short-circuiting of the cells. The dry elements of water activated batteries are stored in a sealed container and are typically capable of indefinite storage life.

Practical, water-activated battery systems have been developed using magnesium anodes in combination with silver chloride or cuprous chloride cathodes. (See Table I below.) In recent years, requirements for specific energy characteristics of reserve current sources have been growing rapidly. Some commercially available water activated current sources are produced based on the following electro-chemical systems: silver chloride-magnesium;

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copper iodide, sulphur-magnesium; and copper chloride-magnesium. Some of these batteries have a specific energy up to 80 W·hr/Kg. In order to improve performance characteristics of water activated reserve batteries, an important problem of utilizing new solid phase oxidizers must be overcome to ensure a high specific energy and the replacement of high cost compounds such as silver-chloride.

TABLE I

	Cathode*	PbCl ₂	Cu ₂ I ₂	CuSCN	CuCl
10	Anode	Mg	Mg	Mg	Mg
	Open Circuit Voltage OCV (V)	1.2	1.5	1.5	1.5
15	Cathode Specific Capacitance (A·hr/g) (Theoretical)	0.193	0.141	0.220	0.271
	Utilization Factor, % of Theoretical	60-75	60-75	60-75	60-75
20	Specific Energy (W·hr/Kg)	50-80	50-80	50-80	50-80
	Volumetric Specific Energy (W·hr/l)	50-120	50-120	50-120	20-150

*Although a silver chloride cathode offers improved performance as compared to other cathode materials, cost factors have mandated the use of the latter.

Anode material consists of a variety of magnesium alloys as described in Table II below (the balance being magnesium):

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TABLE II

	AZ 31		AZ 61		AP 65		MTA-75	
Element	Min. %	Max. %	Min. %	Max. %	Min. %	Max. %	Min. %	Max. %
Al	2.5	3.5	5.8	7.2	6.0	6.7	4.6	5.6
Zn	0.6	1.4	0.4	1.5	0.4	1.5		0.3
Pb					4.4	5.0		
Ti							6.6	7.6
Mn	0.15	0.7	0.15	0.25	0.15	0.30		0.25

AZ 61 is preferred because it tends to sludge and polarize less. AZ 31 gives slightly lower voltage, polarizes at high current densities and sludges more.

Typical cathode materials are as described below:

TABLE III

%Weight				
	Cu ₂ I ₂	CuSCN	PbCl ₂	CuCl
Material	73	75-80	80.7-82.5	95-100
Sulfur	20	10-12		
Additive		0-4	2.3-4.4	
Carbon	7	7-10	9.6-9.8	
Binder		0-2	1.5-1.6	0-5
Wax			3.8	

A cupric oxalate-magnesium sea water cell was disclosed by Hiroi in the *Journal of Applied Electrochemistry*, Vol. 11, pages 589-594 (1981). Hiroi discloses cells with cupric oxalate cathodes bonded with polystyrene and magnesium alloy anodes that are discharged at room temperature. The cathode current efficiency was approximately 75% to a 0.8 V cut-off at an apparent current density of 10 mA/cm². Hiroi also found that cells with sulphur-cupric oxalate cathodes showed discharge performance at higher

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voltages with higher efficiencies than those without sulfur. The drawback of the Hiroi battery is low specific energy, e.g., 235 W·hr/Kg (without structural materials mass).

5 Kegelman, U.S. Patent No. 3,639,174, discloses a voltaic cell which uses a lithium aluminum alloy anode and a copper compound cathode. However, Kegelman teaches the use of a non-aqueous solvent electrolyte.

10 Senderoff, U.S. Patent No. 3,751,298, discloses a thermal-rechargeable electrochemical cell that uses a lithium monoaluminide anode and a cathode of any suitable material including copper chloride. The Senderoff device is rechargeable and uses a lithium tetrachloroaluminate electrolyte.

15 U.S. Patent No. 4,192,913 to Koontz describes a battery having a magnesium based anode, a copper (II) rhodanide-based cathode containing an oxidizer mixed with sulfur and an electroconductive additive. A drawback of this battery is also low specific energy, e.g., 173 W·hr/Kg (without structural materials mass).

It is therefore an object of the present invention to provide a water activated current source that has an improved specific energy level over those available from prior art current sources.

20 It is another object of the present invention to provide a water activated chemical current source that has design simplicity, convenience in operation, and practically unlimited shelf-life before operation.

It is a further object of the present invention to provide a water activated chemical current source that is non-rechargeable and can be used for energizing rescue floating means, emergency radio beacons and the like.

25 It is yet another object of the present invention to provide a water activated chemical current source that utilizes an anode made of magnesium or aluminum or their alloys, a cathode made of a compound consisting of copper hydroxide chloride, sulfur and an electroconductive additive, and an electrolyte of fresh or sea water.

30 It is still another object of the present invention to provide a method of producing a water activated chemical current source having a high specific energy with the use of inexpensive initial materials.

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Summary of Invention

The present invention provides a water activated chemical current source that comprises an anode made of magnesium or aluminum or their alloys, and a cathode of a compound of copper hydroxide chloride, sulfur and an electroconductive additive.

More precisely, the present invention provides a water-activated chemical current source preferably comprising: a magnesium or aluminum based anode; a copper based cathode comprising between about 55 wt. % to about 85 wt. % of copper hydroxide chloride, between about 5 wt. % to about 20 wt. % of sulfur, and between about 1 wt % and about 25 wt. % of an electroconductive additive. A battery in accordance with the present invention is activated by an electrolyte such as a fresh or sea water electrolyte and has a high specific energy as a result of the high electrochemical activity of copper hydroxide chloride. This specific energy is higher than all those known by the inventors and presently commercially available. It is believed that the higher specific power results from higher electrochemical activity of the cathode.

In a method embodiment of the invention, a magnesium-based or aluminum-based anode and a copper-based cathode as described above are provided. The anode and cathode are positioned in spaced relationship in a container adapted to receive a predetermined amount of a water-based electrolyte such that the current producing device can be activated.

These and other objects of the present invention will become apparent in light of the following figures and descriptions.

Brief Description of the Figures

FIG. 1 is a diagram of a battery in accordance with the present invention;

FIG. 2 is a diagram of a preferred embodiment of a water activated chemical current source in accordance with the present invention; and

FIG. 3 is a graph showing a characteristic discharge vs. time curve of one embodiment of the water-activated chemical current source of the present invention.

Detailed Description of the Preferred Embodiment

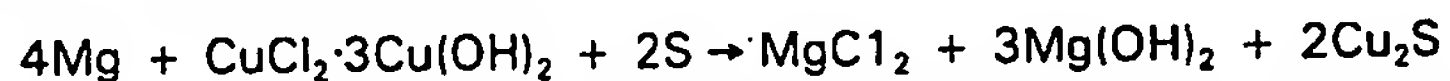
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A preferred embodiment of the present invention can be carried out by using copper hydroxide chloride $[\text{CuCl}_2 \cdot 3\text{Cu}(\text{OH})_2]$ as an oxidizer in a water-activated chemical current source containing a magnesium- or aluminum-based anode. (As used herein, the term "based" when used with a metal denotes the metal or an alloy of the metal.) Exemplary anode materials include alloys selected from the group consisting of AP65, AT61A, AT61B, AZ61, AZ61A, AZ31, Al-Zn (6 wt % Zn), MA2-1 and MTA 75.

In the cathode composition copper hydroxide chloride is present in an amount preferably from about 55 wt % to about 85 wt %. Sulfur is present preferably in an amount from about 5 wt % to about 20 wt %. An electroconductive additive is present in an amount from about 1 wt % to about 25 wt %. Use of the ingredients in proportions outside the ranges set forth above leads to a reduction in the specific energy performance, however the battery is still functional. To improve the suitability of the cathode to pressing and rolling, a binder, such as fluoroplastic or others, can be used in the amount up to 10% relative to the cathode weight.

Copper hydroxide chloride is commercially available, for example from Ural Chemreactive, Verhnyaya Pyshma, Russia (production standard TY6-09-05-730-77), and can be prepared using known processes. Sulfur is also available commercially. Commercially produced elemental sulfur (α -phase) is preferred. Graphite, carbon black, acetylene black, or other carbon compounds are suitable electroconductive additives.

It was discovered that the advantages of the present invention are achieved by the electrochemical system of $\text{CuCl}_2 \cdot 3\text{Cu}(\text{OH})_2$ -Mg. The overall current-forming reaction in the presence of sulfur and an electroconductive additive, for instance, carbon, is:



During this reaction, electrochemical reduction of copper hydroxide chloride having a small electrochemical equivalent equal to 1.992 g/A·hr is performed at potentials of -0.03 to -0.05 V relative to the normal hydrogen electrode potential at current densities of 0.001 to 0.025 A/cm² and with a high oxidizer "utilization factor" 0.75 to 0.9. Here, the term "utilization factor" is used to refer to the fraction of the total oxidizer that is usable to produce energy, with the remaining fraction of oxidizer being unavailable for a variety of reasons. The electrochemical equivalent of copper hydroxide

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chloride (1.992 g/A·hr) compares favorably to that of presently employed materials such as copper oxalate (2.82 g/A·hr). Due to that fact and due to the high utilization factor of copper chloride hydroxide, the battery weight and size can be significantly reduced. This enables one to achieve a specific energy up to 200 W·hr/Kg, depending on the battery mass and dimensions.

In a preferred embodiment, a water-activated chemical current source includes an anode made of magnesium alloy MA2-1, a cathode made of copper hydroxide chloride (71.6 wt. %), sulfur (15.8 wt. %) and carbon (12.6 wt. %), and an electrolyte of a 3% water solution of NaCl.

A cell incorporating this current source has the following characteristics:

	open circuit voltage (V)	1.4
	current density (A/cm ²)	0.005
	average discharge voltage (V)	1.25
	time of discharge (hr)	24
15	specific energy (W·hr/Kg)	324

The time of discharge is defined as the time required for a discharge of the cell to 0.9 V.

The specific energy of the cell is calculated based on the anode and cathode mass, without including the mass of construction materials such as casing, leads, etc., according to the formula:

$$W\cdot hr/Kg = (V_{mean} \cdot C)/m,$$

where V_{mean} is the mean discharge voltage (V), C is the capacity (A·hr), and m is the mass of the anode and cathode (Kg).

For the case of an Al - based anode, for instance Al-Zn (6 wt. % Zn), a device comprising a cathode of the same composition as discussed above and the same electrolyte as discussed above has the following characteristics:

	open circuit voltage (V)	1.35
	current density (A/cm ²)	0.005
	average discharge voltage (V)	1.15
30	time of discharge (hr)	22
	specific energy (W·hr/Kg)	320

Other aluminum alloys can be used provided that their electrochemical potential is close to that of magnesium.

A preferred embodiment of a water activated chemical current source of the invention (Fig. 1) is given as follows:

a) Anode comprising alloy MA2-1 (composition, wt%: Al - 3.8-5.0; Zn - 0.8-1.5; Mn - 0.3-0.7, the balance Mg); thickness 0.5 and 0.8 mm;

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dimensions 35 x 40 mm; number of anodes in a cell, 3 pcs.: one central anode (Fig. 1, reference number 32) 0.8 mm thick and two anodes (Fig. 1, reference number 14) 0.5 mm thick.

b) Cathode comprising copper hydroxide chloride (71.6 wt. %), sulfur (15.8 wt. %), carbon (12.6 wt. %) as graphite powder (6.3%) and carbon black (6.3%); dimensions 35 x 40 x 3.5 mm; number of cathodes in a cell, 2 pcs. To facilitate forming of the cathode by pressing, a fluoroplastic binder was used in the amount of 3% relative to the total weight of the substances comprising the cathode.

c) Separator comprising lignin-based paper impregnated with KCl solution).

The characteristics of the water activated battery are as follows:

Number of cells in battery:	2 pcs.
Mass of battery without electrolyte (Kg)	0.055
Battery casing:	polyethylene
Overall dimensions (mm)	70x48x20
Electrolyte	fresh or sea water.

Testing revealed the following characteristics:

	fresh water	sea water
discharge voltage (V)		
Max	3.0	3.5
Min	1.8	1.8
discharge current, mean (A)	0.12	0.13
period of uninterrupted discharge (hr)	20	20
nominal discharge capacity (A·hr)	2.4	2.6
power (W)	0.26	0.33
specific energy (W·hr/Kg)	95	120

The nominal discharge capacity is the battery capacity at mean discharge current of the battery. It is defined as the product of the mean discharge current and the period of uninterrupted discharge.

In a typical embodiment, the battery includes two elements connected in series within a hermetically sealed polymer case. The case typically has a plurality of holes, such as two holes, for filling with the electrolyte, such as sea water or fresh water. Hydrogen gas and other gaseous by-products of the electrochemical reaction evolved during the operation of the battery are removed through these holes.

Turning now to the drawings, in FIG. 1 a battery 10 of the invention includes cathodes 12 having the above-described composition, i.e.,

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comprising between about 55 wt % and about 85 wt % of copper hydroxide chloride, between about 5 wt % and about 20 wt % sulfur, and between about 1 wt % and about 25 wt % of carbon, and magnesium- or aluminum-based anodes 14 and 32, between which are disposed separators 16 that can
 5 comprise, for example, a lignin-based paper saturated with KCl solution. Dielectric films 18, which may, for example, be polyethylene, are disposed as shown. Positive current lead 20 and negative current lead 22 are connected by connection wires 24 to cathodes 12 and anodes 14 and 32 as shown. Battery 10 further includes case 26 and cover 28, as well as plugs 30 which
 10 plug the holes through which the selected water-based electrolyte is added.

A preferred embodiment of a water activated chemical current source of the invention (Fig 2) is given as follows:

- 15 a) Anode comprising alloy AZ61 (composition, wt%: Al-5.8-7.2; Zn-0.4-1.5; Mn-0.15-0.25, the balance Mg) thickness 1.5mm; dimensions 20mm x 70mm; number of anodes in a cell, 2 pcs (Fig 2. reference number 32).
- b) Cathode comprising copper hydroxide chloride (80.3 wt%), sulfur
 20 (14.2 wt%), carbon (5.0 wt%), elastomer binder (0.5 wt%); dimensions 20mm x 70mm x 3mm, 1 pcs (fig 2, reference number 12).

The characteristics of the water activated cell are as follows:

25	Mass of cell without electrolyte (Kg)	0.032
	Cell casing	polypropylene
	Overall dimensions (mm)	25 x 84 x 12
	Electrolyte	sea water
30	Testing revealed the following characteristics (Fig. 3):	
		sea water
	Discharge voltage (V)	
	Max	1.35
35	Min	0.9
	Discharge current, mean (A)	0.07
	Period of uninterrupted discharge (hr)	50
	Nominal discharge capacity (Ahr)	3.5
	Power (w)	0.09
40	Specific energy (w·hr/kg)	150

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It is specifically contemplated that the present invention can further be practiced utilizing other conventional configurations for water activated batteries.

FIG. 3 shows a typical discharge curve for a cell in accordance with the present invention. The curve illustrated in FIG. 3 was established by
5 discharging an electrochemical cell in accordance with the present invention under simulated operational conditions. To generate this curve, a constant current load was placed across the terminals of the cell. This type of discharge operation was also used to determine operating characteristics of
10 the battery, discussed herein and listed in Table IV, below.

To illustrate the advantages of the present invention, Table IV shows that the characteristics achieved by the water activated current source of the present invention are superior to other sources in the prior art. Each of the cells listed in the table has a construction which is generally similar to
15 embodiments of the present invention. For each of the cells shown in the Table, the cathode composition is indicated in parentheses under the identification of the producer, and the anode composition is indicated below the cathode identification.

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TABLE IV
CHARACTERISTICS OF WATER ACTIVATED CELLS

Type of cell	Producer	Power, (W)	Period of discharge, (hr)	Overall dimensions, (mm)	Mass, (g)	Specific energy (Wh/kg)
L8-1	McMurdo (AgCl) * Anode-Mg	0.30	8	93x26x12	34	70
M8-M	McMurdo (AgCl) * Anode-Mg	0.165	20	93x26x12	34	60
L12B	McMurdo (AgCl) * Anode-Mg	0.25	24	110x30x32	80	75
C1M120	SAFT (Cu, S), Anode-Mg	0.20	12	93x26x12	35	70
Majachok-1	Russian (CuCl) Anode-Mg	0.25	11	115x48x21	85	33
Majachok-2	Russian (CuCl) Anode-Mg	0.25	16	114x61x31	160	25
Present invention		0.33	20	70x48x20	55	120
		0.09	50	25x80x12	32	150

*High cost

Testing of the water activated current source was carried out in approximately 5.5 gallons of fresh and sea water. This amount is sufficient to simulate natural conditions. Small plugs (see FIG. 1) in the device were removed and the device was immersed in water and vigorously swished around to clear any air pockets. The battery was allowed to set for 30 minutes, and then load was attached to the battery to draw a constant current throughout discharge. The device was discharged without interruption for two hours. After two hours, the device was swished around in an attempt to clear internally contained reaction by-products. Periodic jiggling up and down was then continuously applied to simulate both natural

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water wave/current movement and/or movement of the wearer of some type of emergency location device being powered by the battery.

Devices typically operated by a water activated current source include, without limitation: high intensity lamp bulbs with a built-in thermal flasher; 5 slow flashing xenon strobe lamps with an appropriate high voltage step-up transformer, rectifier, storage capacitor, and solid state timer/triggering circuitry; small intermittent radio beacon; low power UHF, two-way transmitter/locator beacon; fishing lures; rescue floating means; meteorological probe; or any device using a battery and having access to a 10 water source. Such devices are rather insensitive to voltage variations like those seen in the illustrated device discharge curve.

Tables V and VI compare the compositions of the anodes, and cathodes of batteries based on copper chloride and copperhydroxide chloride.

Various comparisons have been made which demonstrate the superior 15 characteristics of the present invention. (Table VII).

TABLE V

ANODE	AZ 31		AZ 61	
	Min. %	Max. %	Min. %	Max. %
Al	2.5	3.5	5.8	7.2
Zn	0.6	1.4	0.4	1.5
Mn	0.15	0.7	0.15	0.25

TABLE VI

% / Weight		
Cathode	CuCl	$\text{CuCl}_2 \cdot 3\text{Cu}(\text{OH})_2$
Material	95-100	80.3
Sulfur		14.2
Carbon		5.0
Binder	0-5	0.5

TABLE VII
Performance characteristics

	CuCl	$\text{CuCl}_2 \cdot 3\text{Cu}(\text{OH})_2$
OCV (V)	1.5	1.4
Specific Energy (Wh/Kg)	50-70	150

The present invention thus provides a water activated battery which is a non-rechargeable cell useful for the feeding of lighting apparatus and rescue floating means. The advantages of the invention include complete independence, high specific energy, design simplicity and convenience in operation, a practically unlimited store-life before operation at temperatures from -50 to +50°C, and relatively low cost. Serviceability in sea water and fresh water is afforded within about 0.5-1 min after electrolyte filling.

While this invention has been described in an illustrative manner, it should be understood that the terminology used is intended to be in the nature of words of description rather than of limitation.

Furthermore, while the invention has been described in terms of preferred embodiments thereof, it is to be appreciated that those skilled in the art will readily apply these teachings to other possible variations of the invention.

The embodiments of the invention in which an exclusive property or privilege is claimed are defined as follows:

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What is claimed is:

1. A water activated chemical current source adapted for use with a water-based electrolyte comprising:
 - (i) a magnesium-based or aluminum-based anode; and
 - (ii) a copper-based cathode comprising:
 - (a) between about 55 wt % and about 85 wt % of copper hydroxide chloride,
 - (b) between about 5 wt % and about 20 wt % of sulfur, and
 - (c) between about 1 wt % and about 25 wt % of an electroconductive additive.
2. A water activated chemical current source according to claim 1 wherein said electroconductive additive is selected from the group consisting of carbon, graphite, carbon black, and acetylene black.
3. A water activated chemical current source according to claim 2 wherein said additive is graphite.
4. A water activated chemical current source according to claim 1 wherein said anode comprises an alloy selected from the group consisting of MA2-1, AZ 61, AZ 31, Al-Zn (6 wt % Zn), AZ 61A, AP 65, AT 61A, AT 61B, MTA 75.
5. A water activated chemical current source according to claim 1 wherein said copper-based cathode comprises
 - (a) about 71.6 wt % of copper hydroxide chloride,
 - (b) about 15.8 wt % of sulfur, and
 - (c) about 12.6 wt % of an electroconductive additive.
6. A water activated chemical current source according to claim 1 wherein said cathode consists essentially of the recited components.
7. A water activated chemical current source as claimed in claim 1, further comprising a fresh water or sea water electrolyte.

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8. A water activated chemical current source comprising:
- (i) an anode comprising an alloy selected from the group consisting of MA2-1, AZ 61, AZ 31, Al-Zn (6 wt % Zn), AZ 61A, AP 65, AT 61A, AT 61B, MTA 75, and
 - 5 (ii) a copper-based cathode comprising
 - (a) about 71.6 wt % of copper hydroxide chloride,
 - (b) about 15.8 wt % of sulfur, and
 - (c) about 12.6 wt % of carbon.
9. A method of making a water activated current producing device comprising the steps of:
- (i) providing a magnesium-based or aluminum-based anode;
 - (ii) providing a copper-based cathode comprising
 - 5 (a) between about 55 wt % and about 85 wt % of copper hydroxide chloride,
 - (b) between about 5 wt % and about 20 wt % of sulfur, and
 - (c) between about 1 wt % and about 25 wt % of an electroconductive additive; and
 - 10 (iii) positioning said anode and said cathode in spaced relationship in a container adapted to receive a predetermined amount of a water-based electrolyte such that said current producing device can be activated.
10. A method according to claim 9 wherein said electroconductive additive is selected from the group consisting of carbon, graphite, carbon black, and acetylene black.
11. A method according to claim 10 wherein said electroconductive additive is graphite.
12. A method according to claim 9 wherein said anode comprises an alloy selected from the group consisting of MA2-1, AZ 61, AZ 31, Al-Zn (6 wt % Zn), AZ 61A, AP 65, AT 61A, AT 61B, MTA 75.

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13. A method according to claim 9 wherein said copper-based cathode comprises

- (a) about 71.6 wt % of copper hydroxide chloride,
- (b) about 15.8 wt % of sulfur, and
- 5 (c) about 12.6 wt % of an electroconductive additive.

14. A method as claimed in claim 9 wherein said electrolyte is a fresh water or sea water electrolyte.

15. A water activated chemical current source adapted for use with a water base electrolyte comprising an anode made of a material selected from groups consisting of magnesium, aluminum, or their alloys or mixture thereof; and a cathode comprising copper hydroxide chloride, sulphur and an electro
5 conducting additive.

16. A method of making a water activated current producing device comprising the steps of:

- (i) providing a magnesium-based or aluminum-based anode;
- (ii) providing a copper-based cathode comprising copper hydroxide
5 chloride, sulfur an electroconductive additive; and
- (iii) positioning said anode and said cathode in spaced relationship in a container adapted to receive a predetermined amount of a water-based electrolyte such that said current producing device can be activated.

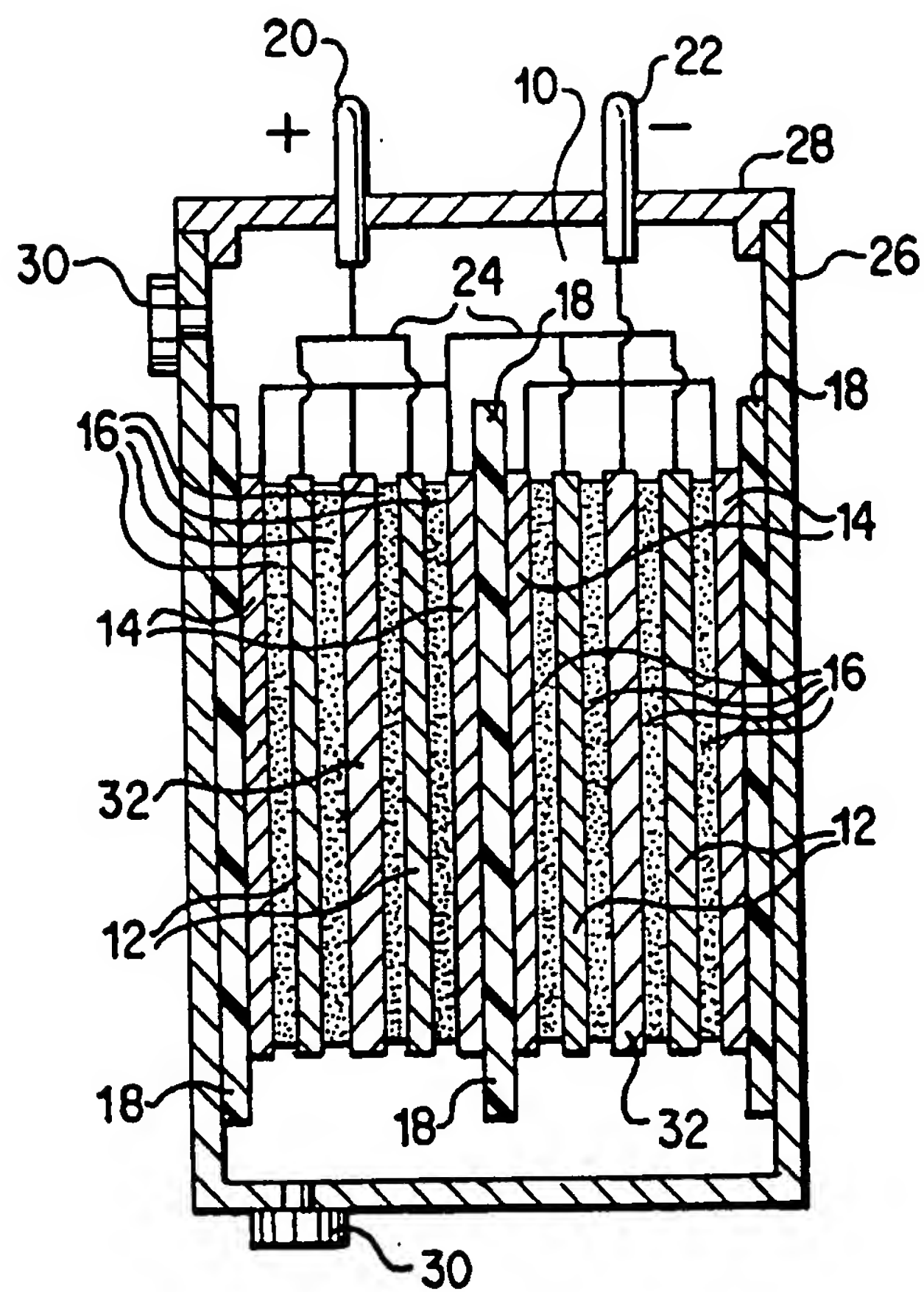


FIG. 1

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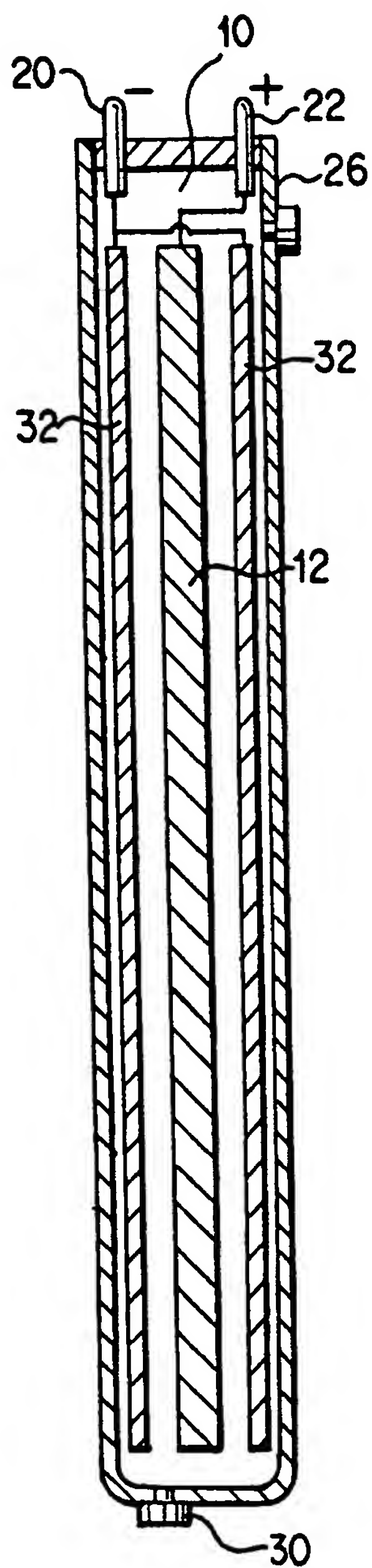


FIG. 2

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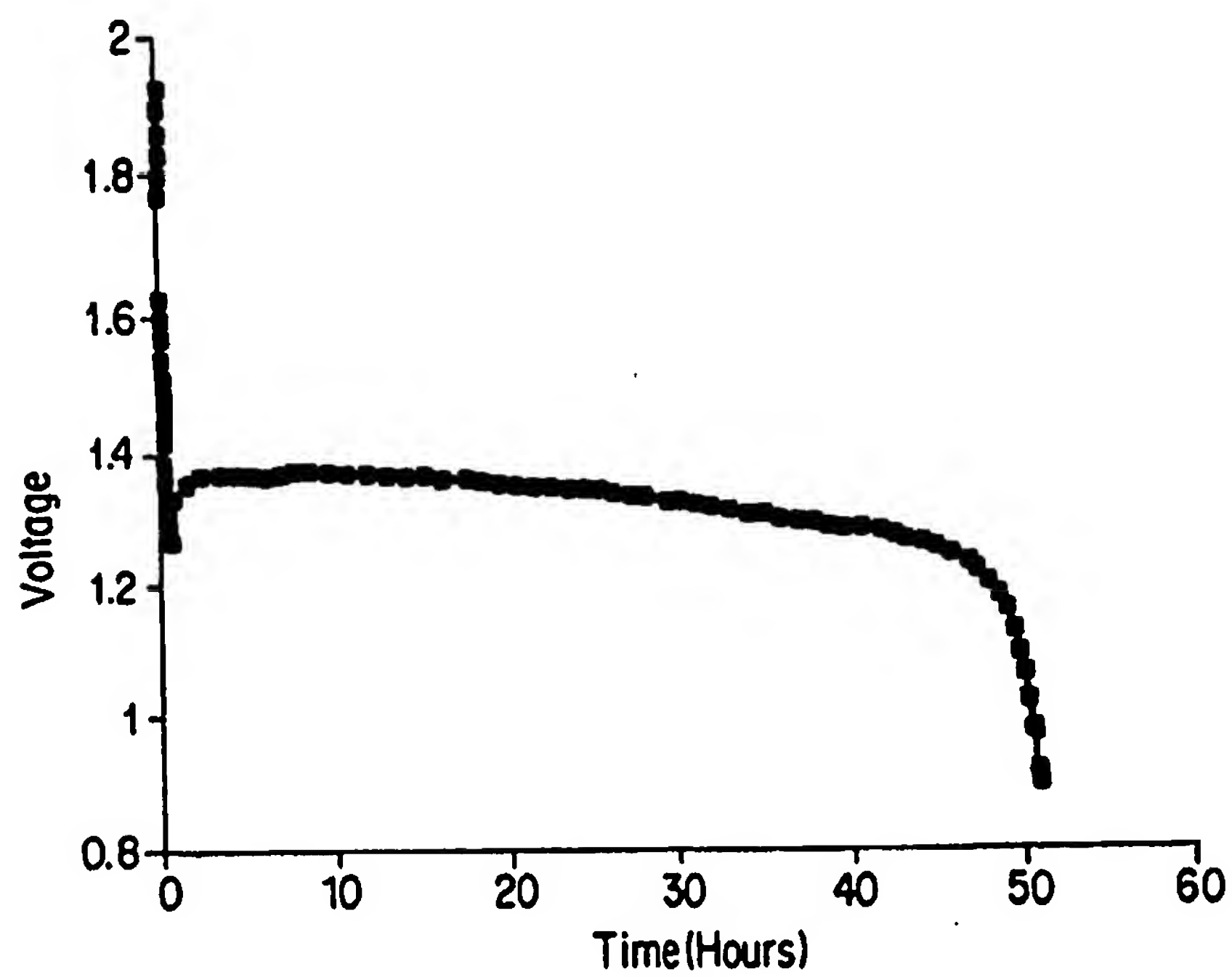


FIG. 3